

## WOOD FILLED COMPOSITES

### ***Technical Field***

This invention relates generally to wood-filled thermoplastic composites preferably polyolefins such as high density polyethylene, medium density polyethylene, low density polyethylene, polypropylene as well as polyvinyl chloride in combination with a cellulose-based filler material for use in the decking industry as synthetic wood for example.

### ***Background of the Invention***

In recent years, extruded cellulose-filled thermoplastic materials have been used in many applications, including window and door manufacture as well as decking material as an outlet for plastic scrap. The use of these wood-filled composites is also growing rapidly, as consumers experience the advantages over wood which include low or no routine maintenance and no cracking, warping or splintering. Additive use is also growing as wood-plastic composites penetrate new markets with more stringent performance requirements and as interest in the long-term stability of composite products increases.

It is known in the art to combine different forms of plastic with different forms of natural fibers or flours, non-limiting illustrative examples including wood flour, crushed shells of nuts, kenaf, hemp, jute, sisal, flax and rice hulls and other natural materials. The purpose of such previous combinations has been to enhance the physical properties and lower the cost of the product. However, such materials have not been successfully used in the form of a structural member that is a direct replacement for wood. Typical common extruded thermoplastic materials have been found not to provide equivalent or acceptable structural properties similar to wood or other traditional structural materials. Accordingly, a substantial need exists for a composite material that can be made of polymer and wood fiber and/or wood flour with an optional, intentional recycle of a waste stream. A further need exists for a composite material that can be extruded into a shape that is a direct substitute for the equivalent milled shape in a wooden or metal structural member. This need requires a material that can be extruded into reproducible stable dimensions, a high compressive strength, an improved resistance to insect attack and rot while in use, and a hardness and rigidity that permits sawing, milling and fastening retention comparable to wood.

Further, companies manufacturing wood-based products have become significantly sensitive to waste streams produced in the manufacture of such products. Substantial quantities of wood waste, including wood trim pieces, sawdust, wood milling by-products,

1 recycled thermoplastic including recycled polyvinyl chloride, have caused significant  
2 expense to various manufacturers. Commonly, these materials are either burned for their  
3 heat value in electrical generation, or are shipped to qualified landfills for disposal. Such  
4 waste streams are contaminated with substantial proportions of hot melt and solvent-based  
5 adhesives, waste thermoplastic such as polyvinyl chloride, paint, preservatives, and other  
6 organic materials. A substantial need exists to find a productive, environmentally  
7 compatible process for using such waste streams for useful structural members and thus,  
8 to avoid returning the materials into the environment in an environmentally harmful way.

9 Therefore, the prior art teaches that conventional structural member applications  
10 have commonly used wood, metal and thermoplastic composites or a combination thereof.

11 The present invention relates to a new and improved process and composition which  
12 provides intimate contact of the wood flour to the plastic matrix, improved dimensional  
13 integrity of the composite, and decreased melt viscosity during processing. The invention  
14 improves over the use of traditional coupling agents which are typically maleic anhydride  
15 grafted polymers, in which the functional group bonds to the more polar wood fibers.  
16 However, the benefit of using this class of coupling agents has not been generally realized  
17 due to its cost.

### 18 ***Summary of the Invention***

19 Accordingly it is a principal object of the invention to provide an alternative to  
20 existing coupling agents which simultaneously provides: lubrication (it contains both internal  
21 and external lubricant systems) with a lower viscosity of wood flour and resin at processing  
22 temperatures; surfactant capability in that it provides a wetting out of the wood flour for  
23 intimate contact of the wood flour to polymer; and superior adhesion in that the internal  
24 bond strength of the overall composite is improved.

25 It is an object of this invention to use chlorinated paraffin waxes such as Chlorez® as  
26 the coupling agent to reduce moisture absorption of the composite, reduce swelling,  
27 improve adhesion as well as improve internal bond strength in addition to acting as a  
28 processing aid.

29 It is another object of this invention to use coupling additives as processing aids in  
30 conjunction with other lubricants, e.g., ethylene bis-stearamide, stearate esters or fatty acid  
31 esters, etc., to increase the bond strength and improve processing of wood-filled  
32 composites in a single package sold commercially under the name Doverbond®.

1 It is still another object of this invention to use Doverbond® formulations to achieve a  
2 much lower extruder torque than comparative examples without Doverbond®.

3 It is still yet another object of this invention to show the use of Doverbond®  
4 formulations wherein the Doverbond® formulation acts both as an internal wetting  
5 (compatibilizer) agent as well as a flow enhancer.

6 It is a further object of this invention to demonstrate the use of Doverbond®  
7 formulations which give higher internal strength values as measured by greater flex  
8 modulus.

9 These and other objects of the present invention will become more readily apparent  
10 from a reading of the following detailed description taken in conjunction with the  
11 accompanying drawings and with further reference to the appended claims.

### 12 ***Brief Description of the Drawings***

13 The invention may take physical form in certain parts and arrangements of parts, a  
14 preferred embodiment of which will be described in detail in the specification and illustrated  
15 in the accompanying drawings which form a part hereof, and wherein:

16 **FIG. 1** is a rheology comparison at 190°C bargraph of Torque (mg) measurements  
17 taken at 6 minutes into Brabender® rheology evaluations;

18 **FIG. 2** is a flexural modulus bargraph of the modulus of elasticity (MOE)  
19 measurements (x1000 psi) evaluated on an Instron® 4200, average of five samples;

20 **FIG. 3** is a tensile properties bargraph of tensile stress at maximum load (psi)  
21 evaluated on an Instron® 4200, average of five samples;

22 **FIG. 4** is a torque rheology evaluation of DB4000 on a Brabender® Plasticorder; and

23 **FIG. 5** is a torque rheology evaluation of zinc stearate/ethylene bis-stearamide  
24 (EBS) on a Brabender® Plasticorder.

### 25 ***Detailed Description of the Invention***

26 Referring now to the drawings wherein the showings are for purposes of illustrating  
27 the preferred embodiment of the invention only and not for purposes of limiting the same,  
28 the Figures show a synergistic effect when using chlorinated resins, e.g., Chlorez® and  
29 certain wood flour and/or wood fiber composites in polymeric composite compositions.  
30 This synergy allows for lower processing torque which translates to higher throughput rates  
31 as well as improved final physical properties in a cost-competitive one-package system.  
32 This is very important in that many thermoplastic extruders are running at essentially full

1 capacity. Reducing processing torque increases extruder output without any corresponding  
2 increase in extrusion lines, thereby enabling each line to run more profitably.

3 The primary processing mode of making these composites is extrusion where the  
4 wood fiber or flour is mixed with molten polymer, typically polyolefin or PVC (although other  
5 thermoplastics are envisioned within the scope of this invention) and then extruded. It is  
6 important to have additives in the compound to promote coupling and lubricity. These  
7 coupling and lubricity additives are very important. The polymer/wood fiber and/or flour  
8 blend is extruded at fairly low temperatures of 180°C, due to the heat sensitivity of the  
9 wood fibers or wood flour. Without the use of lubricants or coupling agents, it is difficult to  
10 extrude a smooth composite having good physical properties. The use of coupling agents  
11 and lubricants helps to improve the long term performance of the composite. Use of proper  
12 coupling agents reduces water absorption and helps maintain mechanical properties after  
13 exposure to water. Coupling agents also improve tensile strength, impact strength, and  
14 creep resistance. The goal is to always try and optimize cost performance with additives.  
15 Currently maleated polypropylene or maleated polyethylene are used as coupling agents.  
16 This current invention discloses the use of chlorinated resins as low-cost processing aids.  
17 Unexpectedly, while only increased extruder output was sought, improved internal bond  
18 strength of the composite was also demonstrated.

19 CHLOREZ® is a registered United States trademark of the Dover Chemical  
20 Corporation, and HORDARESIN® (European trademark associated with same family of  
21 products) and is a family of solid resinous chlorinated paraffins which are especially soluble  
22 in aromatic and chlorinated solvents. They have limited or no solubility in lower alcohols,  
23 glycols, glycerins and water. Chlorinated paraffins are chlorinated derivatives of n-alkanes,  
24 having carbon chain lengths ranging from 10 to 38, and a chlorine content ranging from  
25 about 30 to 70-75% (by weight). The products vary in the distribution, possibly type, range  
26 of chain lengths, and in the degree of chlorination. The melting point of chlorinated  
27 paraffins increases with increasing carbon chain length and with increasing chlorine  
28 content. Consequently, at room temperature, chlorinated paraffins range from colorless to  
29 yellowish liquids at about 40% chlorine, to white solids (softening point at about 90°C) at  
30 70% chlorine. Chlorinated paraffins have very low vapor pressures (e.g.,  $1.3 \times 10^{-4}$  Pa for  
31 C<sub>14-17</sub>, 52% Cl at 20°C) and solubilities in water, the latter ranging from 95 to 470  
32 microgram/liter for some of the short chain mixtures (C<sub>10-13</sub>) to as low as 3.6 to 6.6  
33 micrograms/liter for some of the longer chain mixtures (C<sub>20-30</sub>). In a preferred embodiment,

1 the resin will be chlorinated to between approximately 30-75%. In a more preferred  
2 embodiment, the resin will be chlorinated to between approximately 40-75%. In a still more  
3 preferred embodiment, the resin will be chlorinated to between approximately 50-75%. In a  
4 most preferred embodiment, the resin will be chlorinated to between approximately 68-  
5 72%. In this embodiment, the resin will be a solid.

6 Traditional wood-filled composites are comprised of primarily four components: (a)  
7 polymer resin; (b) wood flour or fiber (depending on the mesh size and aspect ratio of the  
8 wood-based filler); (c) lubricant / processing aid; and (d) coupling agents. Optionally, other  
9 additives such as colorants, ultra-violet degradation inhibitors; anti-fungicidal components;  
10 and anti-microbial components are blended into the composite.

11 One of the keys to the functional performance of the coupling agent is to provide  
12 intimate contact of the wood flour with the plastic matrix. It is also used to improve the  
13 dimensional integrity of the composite as well as decrease the melt viscosity during  
14 processing. The most commonly used coupling agents in the Prior Art are maleic  
15 anhydride grafted polymers which are employed as a surfactant, wherein the functional  
16 group bonds are used to bond to the polar wood fiber. These additives are not used  
17 extensively, primarily due to cost, particularly since no economically realized performance  
18 benefit is demonstrated for the increased cost.

19 Through the use of the Doverbond<sup>®</sup> formulations, this coupling agent acts as a  
20 lubricant in that it: contains both internal and external lubricant systems, leading to a  
21 lowered viscosity of the wood flour and resin composite at processing temperatures; acts  
22 as a surfactant, providing a "wetting out" of the wood component for intimate contact  
23 between the wood flour or fiber and polymer; and improves adhesion by providing  
24 improvements in internal bond strength of the overall composite.

25 Experimentally, a 0.55 MFI High Density Polyethylene (HDPE) sold commercially  
26 under the trademark Fortiflex<sup>®</sup> B53-35H-FLK from BP Solvay, was used in the evaluations  
27 and loaded according to **Table 1**. The natural filler is un-dried 40-mesh hardwood Maple  
28 flour from American Wood Fibers, loaded at 60% in all formulations. The experimental  
29 systems were all tested against a standard 1:1 ratio of ethylene bis-stearamide (EBS) wax  
30 and zinc stearate, loaded at 5%, and a control system consisting of 40% HDPE and 60%  
31 Maple flour. Each experimental Doverbond<sup>®</sup> system was run individually, loaded at 5%,  
32 and again with an additional process aid loaded at 3%, see **Table 1**.

**Table 1**

Formula	Standard	DB <sup>(5)</sup> 1000	DB 2000	DB 2300	DB 3000	DB 3300	DB 4000	DB 4300	Control
HDPE	35	35	35	32	35	32	35	32	40
Maple Flour	60	60	60	60	60	60	60	60	60
Chlorez <sup>®</sup>		5	2.5	2.5	2.5	2.5	4	4	
ZnSt/EBS 1/1	5								
Lubricant A <sup>(1)</sup>			2.5	2.5					
Lubricant B <sup>(2)</sup>					2.5	2.5			
Lubricant C <sup>(3)</sup>							1	1	
Process Aid <sup>(4)</sup>				3		3		3	
(%)	100	100	100	100	100	100	100	100	100

<sup>(1)</sup> ethylene bis-stearamide

<sup>(2)</sup> pentaerythritol tetrastearate

<sup>(3)</sup> polyethylene glycol (PEG) monostearate (M.W. = 1500)

<sup>(4)</sup> talc

<sup>(5)</sup> the reference to DB pertains to the additives exclusive of the thermoplastic and cellulose-based filler.

Flexural modulus samples were accurately weighed and mixed by hand according to **Table 1**, in 1100g “batches.” Each sample was compounded in a Banbury<sup>®</sup> mixer set at 180°C for 5 minutes. Each sample was immediately removed and compression molded at 190°C/25,000psi for 5 minutes and cooled for 15 minutes @ 25,000 psi. The size of the finished sample was 6” X 6” X 0.25”. Each sample was then cut into bars measuring 5” X 0.50” X 0.25” for testing. Flexural modulus, or modulus of elasticity (MOE), was measured according to ASTM D-790 Method 1. Tensile properties were measured on Type-I test bars in accordance with ASTM D-638, on samples cut from the previously mentioned compression-molded plaques.

Rheology measurements were performed on 50-gram samples prepared according to **Table 1**. Meter-grams of torque (mg) and temperature (°C) measurements were derived from evaluations performed on a Brabender<sup>®</sup> Plasticorder PL2000 3-zone mixing bowl. Baseline torque measurements were derived from the reading taken at 6 minutes into each evaluation; this was kept constant throughout the study and reported in **FIG. 1**. The mixing bowl temperature was set at 190°C and at a speed of 60 rpm; the samples ran for 20

1 minutes each before the test was terminated. The tested sample was then removed from  
2 the mixing bowl and compression molded into a 3" X 3" plaque at 190°C for 2 minutes to  
3 compare relative heat stability based on color generation.

4 Referring now to the drawings wherein the showings are for the purpose of  
5 illustrating a preferred embodiment of the invention only and not for the purpose of limiting  
6 same, there is shown a significant improvement in final physical properties in cellulose-filled  
7 plastic composites as well as significant improvements in viscosity reduction which results  
8 in improved extruder throughput when Doverbond® is added to the composite.

9 The Doverbond® product is a multi-component one-pack system where an individual  
10 component aids in only one of the above property areas. These property areas are  
11 positively quantified by an increase in flexural modulus, an increase in tensile strength or a  
12 decrease in torque. The most effective one-pack system will then have a positive effect on  
13 all three property areas.

14 DB1000, which is the base coupling agent component for the entire Doverbond®  
15 line, is extremely effective at increasing both the flexural modulus and tensile strength over  
16 that of the standard system, as shown in **FIGS. 2 & 3** respectively. This coupling effect is  
17 demonstrated when the standard system is replaced with DB1000, a 64% increase in  
18 tensile strength and a 34% increase in flexural modulus is resulted. The only drawback  
19 then, is the increase in torque associated with this action.

20 Three different lubricant chemistries were evaluated, Lubricants A, B, and C, as  
21 shown and identified in **Table 1**. The overall additive system loading was kept constant at  
22 5% and the coupling agent and lubricant package ratios were varied.

23 The 1:1 addition of the coupling agent to Lubricant A (DB2000) resulted in an  
24 increase in tensile strength and flexural modulus, but the lubricating effect was not realized,  
25 as shown in **FIG. 1**.

26 The 1:1 addition of the coupling agent to Lubricant B (DB3000) demonstrated similar  
27 results, where flexural modulus was maintained and tensile strength was improved,  
28 compared to the standard system; **FIGS. 2 & 3** respectively.

29 The 4:1 addition of the coupling agent to Lubricant C (DB4000) shows the  
30 effectiveness of this lubricant chemistry. Lubricant C was only loaded at 1% to the overall  
31 formulation, compared to 2.5% of the other lubricants, **Table 1**. The DB4000 system  
32 outperformed the standard formulation in all required categories; the torque was reduced  
33 by 22% (**Figure 1**), flexural modulus was maintained (**FIG. 2**), and the tensile strength was

1 increased by 62% (**FIG. 3**). Another interesting aspect of the DB4000 system is the  
2 improvement in thermal stability. Compare the Brabender chart in **FIG. 4** with that of the  
3 standard formulation in **FIG. 5**. Note the “flat-line” effect with the DB4000, indicative of a  
4 highly stable system, even after running for 20 minutes at 190°C set point. Also worthy of  
5 note is the drop in temperature, which displays the effectiveness of Lubricant C as well,  
6 shown in **FIG. 4**. This temperature drop is typically due to the reduction of shear forces  
7 associated with processing. The effect of temperature reduction coupled with the drop in  
8 torque throughout the entire test is exhibited in the pressed plaques of the actual tested  
9 samples.

10 Another series of tests were performed where a process aid was added in addition  
11 to the previously outlined formulations; see **Table 1**.

12 The addition of the process aid to the system containing Lubricant A showed a  
13 positive synergy where torque was significantly reduced, **FIG. 1**, and tensile properties  
14 were increased, **FIG. 2**. Flexural modulus was not greatly affected.

15 This synergy was noticed more in conjunction with Lubricant B where all three  
16 important categories showed an improvement over the DB3000 system. Comparing to the  
17 standard formulation, DB3300 showed a 27% increase in flexural modulus (**FIG. 2**) and a  
18 50% increase in tensile strength (**FIG. 3**). The melt viscosity remained at a 27% increase  
19 over standard.

20 The addition of the process aid to Lubricant C (DB4300) displayed marked  
21 improvements in all categories when compared to the industry standard formulation.  
22 DB4300 presents a system that can offer a 36% reduction in torque (**FIG. 1**), a 9% increase  
23 in flexural modulus (**FIG. 2**), and a 52% increase in tensile strength as seen in **FIG. 3**. A  
24 similar effect, as previously discussed, was also noticed where the color retention of the  
25 tested sample was improved. Therefore, one-pack systems can be designed to incorporate  
26 coupling agents, lubricants, and process aids, which result in improved mechanical  
27 properties and potentially better flow rates.

28 While chlorinated resins are believed to be the preferred coupling agent, in some  
29 instances, it is desirable to add additional coupling agents, e.g., interfacial agents which aid  
30 with the intimate blending of the dissimilar surfaces of wood flour (hydrophilic) and polymer  
31 (hydrophobic). The interfacial agent acts as a polymeric surfactant and aids in the formation  
32 of the polymer/wood flour blend through its dual functionality of having at least one portion  
33 of the moiety being hydrophilic and at least one other portion of the molecule being



hydrophobic. Perhaps phrased another way, the moiety must be functionalized to the extent wherein at least one part of the molecule can bond either in a chemical or a physical sense, to at least the cellulose component of the wood flour while at least one other portion of the molecule can mix and/or compatibilize with the polymer.

The impact of lower levels of chlorinated resins were analyzed in **Table 2** in which a Brabender® study was run in the bowl at 175°C for 20 minutes. Samples were pulled at 2, 6, 10, and 20 minutes. The color progression of all samples looked the same. All held good color. Banbury® batches were prepared of each formulation, 175°C for 5 minute mixing cycle. Physical properties were measured from test specimens cut from plaques compression molded to 0.25 inch thickness. The formulations, torques, and properties are as follows.

**Table 2**

Formula	Standard	A	B	C	D	E	E	F	G	H	I
HDPE	35	35	32	35	32	35	32	35	35	35	32
Maple Flour	60	60	60	60	60	60	60	60	60	60	60
Chlorez®		2.5	2.5	2.5	2.5	4	4				
CPE <sup>(5)</sup>								5	2.5	2.5	2.5
Zn <sub>2</sub> O	2.5										
Lubricant A <sup>(1)</sup>	2.5	2.5	2.5								
Lubricant B <sup>(2)</sup>				2.5	2.5				2.5		
Lubricant C <sup>(3)</sup>						1	1			2.5	2.5
Process Aid <sup>(4)</sup>			3		3		3				3
(%)	100	100	100	100	100	100	100	100	100	100	100
Torque (6 min)	627	689	590	863	721	799	610	1441	1712	746	640
Tensile (psi)	975	1220	1570	1400	1470	1500	1480	2100	1840	1400	1340
Elongation (%)	0.77	1.1	0.4	0.9	0.76	1.4	0.6	1.0	1.6	1.3	0.5
Flex Modulus (psi x 10 <sup>3</sup> )	257	341	331.6	260	319.8	259.3	281.4	223.3	209.2	167.2	180

<sup>(1)</sup> ethylene bis-stearamide

<sup>(2)</sup> pentaerythritol tetrastearate (PES-125)

<sup>(3)</sup> polyethylene glycol (PEG) monostearate (M.W. = 1500)

<sup>(4)</sup> talc

<sup>(5)</sup> 36% chlorinated polyethylene.

1 It is envisioned that a number of polymers are capable of acting as an interfacial  
2 agent between the cellulose surfaces in the wood flour, which have a high hydroxy content,  
3 and the polymer phase, e.g., polyvinyl chloride. Without being limited to any one theory, it is  
4 believed that the interfacial agent adsorbs on the surface of the cellulose particles and  
5 makes that surface "look" more polymer-like to the surrounding polyvinyl chloride. Hence,  
6 any polymeric compound likely to physisorb or chemisorb on cellulose is believed to  
7 provide the desired interfacial blending necessary to effectively form the desired product  
8 blend.

9 Various copolymers effective in this application would include copolymers of  
10 ethylene and acrylic acid, i.e. poly(ethylene-co-acrylic acid),  $(-CH_2CH_2-)_x [-CH_2CH(CO_2H)-]_y$ , commercially available with varying acrylic acid content. One of the  
11 keys to the efficacy of this group of compounds is the "-co-acrylic acid" or similar type of  
12 polymer grouping. Other promising candidates of this sort would include: poly(ethylene-co-  
13 methacrylic acid), poly(ethylene-co-methyl acrylate-co-acrylic acid), poly(methyl  
14 methacrylate-co-methacrylic acid), and poly(tert-butyl acrylate-co-ethyl acrylate-co-  
15 methacrylic acid).

17 Another characteristic believed to play a role in the efficacy of the interfacial agent is  
18 its hydroxy content. Assuming physisorption is the predominant mechanism, then  
19 compounds which are believed to aid in the composition would include: poly(styrene-co-  
20 allyl alcohol), and poly(vinyl alcohol-co-ethylene).

21 Without being held to any one theory of operation, it is believed that when  
22 chemisorption is at least one of the operative modes of this invention regarding the  
23 interfacial agent and the cellulose, then any carboxylic acid group containing polymer will  
24 have at least some degree of efficacy in this system. Additionally, ester bonds can be  
25 formed from amides, acrylates, acyl halides, nitriles and acid anhydrides reacting with  
26 hydroxyl groups. Additional representative polymers would include: poly(vinyl chloride),  
27 carboxylated, poly(vinyl chloride-co-vinyl acetate-co-maleic anhydride), and various -co-  
28 maleic acid or -graft-maleic acid polymers, of which there are many.

29 Amides will react with alcohols under acidic conditions to produce an ester and an  
30 ammonium salt, rather than water as in the case with carboxylic acids, of which  
31 representative examples would include: polyacrylamide, and poly(acrylamide-co-acrylic  
32 acid), although the hygroscopic qualities of these polymers somewhat diminish their  
33 effectiveness in this application.

Another chemistry which is applicable is that of the acrylates, which are a subset of esters. It would be possible to form an ester bond with an alcohol producing another alcohol in a transesterification reaction. For example, a methacrylate containing polymer could react with the surface hydroxyl to form the surface ester bond and methanol. Representative examples would include: poly(methyl methacrylate), poly(ethyl methacrylate), poly(ethylene-co-ethyl acrylate), and poly(butyl acrylate).

It is also known that acyl halides can react with an hydroxyl group to yield the ester bond and HCl. Another reaction chemistry would include that of a nitrile with a hydroxyl group under acidic conditions to yield the ester bond and an ammonium salt. Representative examples would include: polyacrylonitrile; and poly(acrylonitrile-co-butadiene), particularly when the above poly(acrylonitrile-co-butadiene) is functionalized via amine termination or carboxylation.

Another reaction which is possible is via an acid anhydride which reacts with a hydroxyl group to give the ester bond and an ester. A representative example would include: poly(ethylene-co-ethyl acrylate-co-maleic anhydride).

Another family of block copolymers which are believed to be effective in this composition would be those formed with polyacrylic or polymethacrylic acid, e.g., polystyrene di-block copolymers such as polystyrene-b-polyacrylic acid and polystyrene-b-polymethacrylic acid. Other candidates include block copolymers with polyvinyl alcohol or polyoxyethylene.

Once again, without being limited to any one theory of operation, it is conceivable that any hydroxyl, hydroxy or acid functionalized low to medium molecular weight polymers may serve as compatibilizers in this system, e.g., hydroxyl functionalized polybutadiene [CAS 69102-90-5]. Other compounds which may act similarly would include poly(vinyl chloride-co-vinyl acetate), poly(vinyl chloride-co-vinyl acetate-co-2-hydroxypropyl acrylate), poly(vinyl chloride-co-vinyl acetate-co-maleic acid).

As used in this application, the term cellulose-based is meant to include all types of material containing cellulose, a non-limiting example listing including wood flour, wood fiber, rice hulls, cotton, wool, bamboo, sisal, kenaf, jute, crushed shells of nuts, hemp, flax and other natural materials etc. The targeted mesh size of the cellulose-based filler is dependent upon the end-use application, and both flour and fiber forms of cellulose are envisioned to be applicable. In some embodiments, synthetic fibers may also be used in conjunction with the cellulose-based fibers, e.g., polyester or aramide as well as inorganic

1 fibers (chopped or long), for example, glass fibers, carbon fiber and ceramic fibers. The  
2 amount of cellulose-containing material can vary widely, with ranges from 10-80% by  
3 weight of the molded or extruded articles.

4 Many lubricants are applicable for use in this invention, a non-limiting illustrative list  
5 including: metal soaps, hydrocarbon waxes, fatty acids, long-chain alcohols, fatty acid  
6 esters, particularly esters of long chain ( $C_{16}$  to  $C_{24}$ ) fatty acids with polyalkylene glycols,  
7 fatty acid amides, silicones, fluorochemicals, acrylics, and mixtures thereof. Preferred are  
8 long chain fatty acids (e.g., stearic, oleic, palmitic, lauric, tallow acids, etc.) with  
9 polyalkylene or polyoxyalkylene glycols (e.g., polyethylene glycol, polypropylene glycol,  
10 etc.) to form polyalkylene mono- or di- esters. These preferred lubricants have surfactant  
11 characteristics and are generally nonionic. As general guidance it is preferred that these  
12 lubricants when used in the preparation of formulations of this invention be selected from  
13 those surfactants classified as anionic or nonionic. These surfactants are particularly useful  
14 for their compatibility and stability. Surfactants generally suitable for the various purposes in  
15 the present invention include long chain ( $C_{16}$  to  $C_{24}$ ) fatty acids, e.g. palmitic acid, stearic  
16 acid and oleic acid; esters of long chain ( $C_{16}$  to  $C_{24}$ ) fatty acids, e.g. sodium palmitate,  
17 sodium stearate and sodium oleate; sodium lauryl sulphate; polyethylene glycol;  
18 polyethylene glycol alkyl ethers; fatty acid esters of polyethylene glycol, e.g. polyethylene  
19 glycol mono- or di-stearate; propylene glycol; fatty acid esters of propylene glycol, e.g.  
20 propylene glycol monostearate; glycerine; fatty acid mono- or poly-glycerides, such as  
21 glyceryl monostearate; polyoxyethylene fatty acid esters, ethers and amines, e.g.  
22 polyoxyethylene mono- and di-stearate, and polyoxyethylene lauryl ether; polyoxyethylene  
23 sorbitan esters, e.g. polyoxyethylene sorbitan monolaurate, monopalmitate, monostearate  
24 or mono-oleate; polyoxyethylene alkyl phenols and alkyl phenyl ethers; polyoxyethylene  
25 castor oil; sorbitan fatty acid esters; the polysorbates; stearylamine; triethanolamine oleate;  
26 vegetable oils, e.g. sesame seed oil or corn oil; cholesterol; and tragacanth. The amount of  
27 lubricants is from 0.1% to 20% by weight of the molded or extruded articles, more  
28 preferably 0.1 to 4%, most preferably 1 to 3% by weight.

29 Many thermoplastic resins are applicable in this invention, a non-limiting illustrative  
30 list including polyolefins: such as polypropylene, polyethylene and polybutenes, as well as  
31 diolefins, e.g., polybutadiene and isoprene; acrylonitrile-styrene-butadiene block  
32 copolymers; polystyrene; polyamides such as nylons; polyesters; polyvinyl chloride;  
33 polycarbonates; acryl resins and thermoplastic elastomers such as EPDM (ethylene

propylene diene copolymers), and they are used singly or as a mixture thereof, or as a polymer alloy using them. Among them, polyethylene and polypropylene are preferred.

As illustrated above, while chlorinated paraffin waxes are the preferred coupling agent, many others as discussed previously are applicable to supplement the chlorinated wax base agent, including mixtures thereof. The percent of chlorination in the coupling agent can vary widely, with chlorine contents ranging from about 30% to 70-75%.

Preferably, the chlorinated wax is a solid, most preferably, a paraffin wax sold commercially by the Dover Chemical company under the trademark Chlorez<sup>®</sup> having a chlorine content of about 68-72%. The amount of coupling agent (interfacial bonding agent and/or surfactant) is from 0.1% to 10% by weight of the molded or extruded articles, preferably from 1 to 8%, and more preferably from 3-5%.

When used, the processing aid is a nucleating agent selected from the non-limiting illustrative list of polyhydroxybutyrate, sorbitol acetal, boron nitride, titanium oxide, talc, clay, calcium carbonate, sodium chloride, metal phosphate, and mixtures thereof. The amount of processing aid is from 0.1% to 30% by weight of the molded or extruded articles, typically approximately 10% by weight.

In addition, various kinds of conventionally used stabilizers, pigments and antistatic agents may be compounded as necessary, and depending on the intended use, various kinds of other modifiers for example, surface characteristic modifiers such as gloss agents, antistatic agents and surface processing assistants as well as biological characteristic modifiers such as antimicrobial agents, anti-fungus agents and preservatives may be compounded as necessary.

In the foregoing description, certain terms have been used for brevity, clearness and understanding; but no unnecessary limitations are to be implied therefrom beyond the requirements of the prior art, because such terms are used for descriptive purposes and are intended to be broadly construed. Moreover, the description and illustration of the invention is by way of example, and the scope of the invention is not limited to the exact details shown or described.

This invention has been described in detail with reference to specific embodiments thereof, including the respective best modes for carrying out each embodiment. It shall be understood that these illustrations are by way of example and not by way of limitation.